



WO 9606888A1

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- 1 -

**TITLE****LIQUID CRYSTALLINE POLYMER COMPOSITION****FIELD OF THE INVENTION**

5 The present invention relates to a composition comprising an aromatic liquid crystalline polymer (LCP) and a low molecular weight polyethylene (PE) which acts as a lubricant.

**TECHNICAL BACKGROUND**

10 Thermotropic liquid crystalline polymers are useful as molding resins for a variety of applications. This is due to the desirable properties that many of these polymers possess, such as good moldability, low flammability, high temperature resistance, good physical properties, etc. For electrical applications, such as electrical connectors, low flammability or resistance to burning is important. Further, in many electrical applications, the parts are intricate and have thin walls. As such, these parts are  
15 sometimes difficult to eject from the mold. Internal lubricants (sometimes called mold release agents) are useful to aid in the ejection of such parts from the mold. For a discussion of such lubricants, see H. Mark, et al., Ed., Encyclopedia of Polymer Science and Engineering, Vol. 14, John Wiley & Sons, New York, 1988, p. 411-421.

20 While internal lubricants are generally desirable in many applications, it is preferable that the desired properties of the LCP are degraded as little as possible. It has been found that low molecular weight polyethylene (often called a "wax") acts as a lubricant in LCPs. Surprisingly, even though polyethylene itself burns quite readily, its addition to an LCP  
25 does not significantly degrade the low flammability properties of the LCP.

**SUMMARY OF THE INVENTION**

This invention relates to a composition comprising an aromatic liquid crystalline polymer and from about 0.05 to about 2.5 percent by weight of said aromatic liquid crystalline polymer of polyethylene, said  
30 polyethylene having a molecular weight within the range between about 1,000 to about 10,000.

**DETAILS OF THE INVENTION**

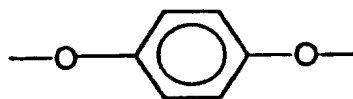
The LCPs useful herein are aromatic LCPs. By an "aromatic" LCP is meant that the backbone of the polymer is composed of aromatic  
35 rings and functional groups such as esters, amides and imides. There are no aliphatic or cycloaliphatic groups which are part of the main chain.

- 2 -

However, such groups may be substituted onto the main chain as side groups. For instance, in an LCP which is at least partially a polyester, repeat units may be derived from t-butylhydroquinone, methylhydroquinone, phenethylhydroquinone, methylterephthalic acid, t-butyl-4-hydroxybenzoic acid, etc.

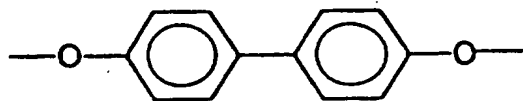
Preferred aromatic LCPs are polyesters, poly(imide-esters) and poly(amide-esters), with polyesters being especially preferred. These types of polymers are well known to the artisan, see for instance U. S. Patents 4,900,804, 4,851,497, 4,067,852, 4,083,832, 4,727,131, 4,727,129, 4,762,907, 4,664,972, 4,118,372, 4,684,712, 4,522,974, 4,473,682 4,161,470 and 5,110,896. Preferred LCPs may contain repeat units derived from one or more of 4-hydroxybenzoic acid, 6-hydroxynapthoic acid, terephthalic acid, isophthalic acid, 2,6-naphthalene dicarboxylic acid, 4,4'-bibenzoic acid, hydroquinone, 4,4-biphenol, resorcinol, 2,6-dihydroxynaphthalene, substituted hydroquinones including chlorohydroquinone, methylhydroquinone, t-butylhydroquinone, phenylhydroquinone, and phenethylhydroquinone. Especially preferred polymers include polymers consisting essentially of

(a) structure (I),



(I)

(b) structure (II),

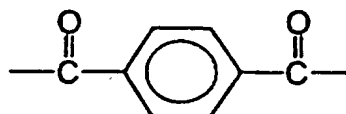


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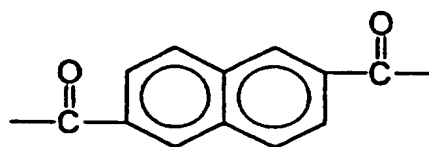
(c) structure (III),



(III)

5

(d) structure (IV),

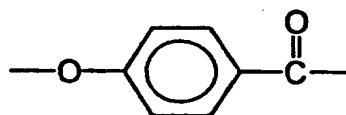


(IV)

10

and

(e) structure (V)



(V)

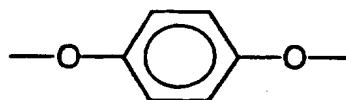
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wherein the molar ratio of (I):(II) ranges from 0:100 to 100:0, wherein the molar ratio of (III):(IV) ranges from 85:15 to 15:85, wherein the molar ratio of the total of (I) and (II) to the total of (III) and (IV) is substantially 1:1, and further wherein there are 100 to 600 moles of (V) per 100 moles of (I) plus (II);

20

or polymers consisting essentially of the following repeat units:

(a) structure (VI)



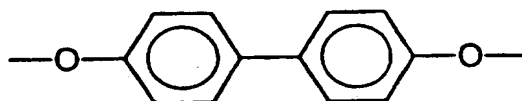
(VI)

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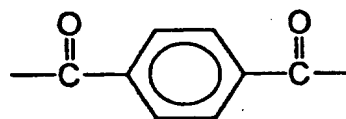
and/or  
structure (VII)



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(VII)

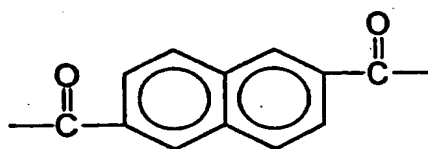
(b) structure (VIII)



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(VIII)

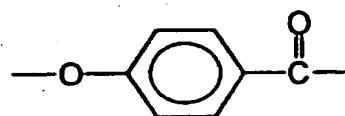
(c) structure (IX)



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(IX)

(d) structure (X)

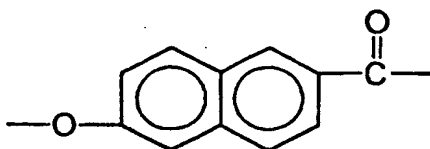


20

(X)

and  
(e) structure (XI)

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(XI)

wherein the molar ratio of (VI):(VII) ranges from 0:100 to 100:0, the molar ratio of (VIII):(IX) ranges from 25:75 to 90:10, the molar ratio of the sum of (VI) and (VII) to the sum of (VIII) and (IX) is substantially 1:1, the molar ratio of (IX):(X) ranges from 97:3 to 50:50, and the number of moles of (IX) plus (X) ranges from 100 to 600 per moles of (VI) plus (VII). Especially preferred polymers also include copolymers of p-hydroxybenzoic acid and 6-hydroxynaphthoic acid.

The PE useful herein has a molecular weight within the range between about 1,000 to about 10,000, preferably about 1,500 to about 4,000. Low pressure polymerized (high density), polar PE is preferred for use in the present invention. Such PEs are made by oxidation of PE which introduces polar functional groups, such as carboxyl groups, into the PE. Such oxidized PEs are commercially available, such as those from Hoechst Celanese Corp., Somerville, NJ, U.S.A. under the designation "Hoechst Wax PED".

The amount of PE present in the present invention generally ranges from about 0.05 to about 2.5 percent by weight of the LCP, preferably about 0.1 to about 2.0 percent by weight, and most preferably about 0.2 to about 1.0 percent by weight. Other materials usually found in LCP molding compositions may also be present, such as fillers, including glass, clay, talc and other minerals, and carbon black, pigments, antioxidants, other polymers, etc. Preferred fillers are glass fiber, talc, titanium dioxide and carbon black, and an especially preferred filler is glass fiber.

Flammability or flame resistance properties reported herein were measured by UL-94, revised as of May 14, 1993, and published by Underwriters Laboratories, Inc., Melville, NY, U.S.A. It is preferred that the flammability (as measured by the total after flame time for any condition set) of the compositions not containing PE is increased by less than 50% when the PE is added, more preferably less than 25%, and especially preferably increased less than 10%. It is also preferred that the overall UL-94 rating under the particular test conditions used is the same or better (less

- 6 -

flammable rating) for the PE containing composition compared to the composition without PE. It is particularly preferred if the PE containing composition has a UL-94 rating of V-0.

5 The PE containing LCP compositions claimed herein may be made by standard methods. For instance these materials (and others to be in the composition) may be mixed together in a single or twin screw extruder. The composition may be then directly melt formed or formed into pellets for later use. Melt forming may be done by extrusion or injection molding, for example, and the latter is preferred. Such formed parts are  
10 useful as films, or as molded parts in the automotive, electronic or aerospace applications. As mentioned above, the composition is particularly useful for electronic parts, particularly electronic connectors.

#### **EXAMPLES 1-3**

15 The LCP used in these Examples had the composition (structures shown above, in mole percent) (I), 9.6%; (II), 9.6%; (III), 13.5%; (IV), 5.8%; and (V), 61.5% (see U.S. Patent 5,110,896). The fiberglass used was Owens-Corning Fiberglass Corp., grade 408. The polyethylene was reported to be an oxidized low pressure polyethylene and was obtained from Hoechst Celanese Corp., Somerville, NJ, U.S.A. as grade PED 521. It reportedly had  
20 a drop point of about 105°C, an acid value of about 35 mg KOH/g and a density of about 0.95 at 20°C, using test methods specified by Hoechst Celanese Corp.

The materials were mixed on a Werner and Pfleiderer 28 mm twin screw extruder, in which the zone temperatures (rear to front) were  
25 approximately were 255°C, 290°C, 290°C, 290°C, 300°C and 345°C (die). A one hole die was used, and pellets were made. The pellets were then injection molded into test bars, and temperatures in the injection molding machine (rear to front) were approximately 325°C, 320°C, 330°C and 335°C-347°C.

30 Compositions (weight percents are based on the total amount of everything in the LCP compositions), tensile strengths and elongations, and UL-94 burn times and overall ratings are given in Table 1, along with values for Comparative Example A, which contained no PE. Tensile properties were determined with bars that were 0.318 mm (1/8") thick, while the UL-94

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- 7 -

tests were done with bars that were 0.079 mm (1/32") thick, with the burns being done on dry, as-molded, bars. Tensile tests were performed by the procedure of ASTM D638.

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TABLE 1

Ex. No.	wt % PE	wt % Fiber-glass	Tensile Strength (MPa)	Elong. %	Burn Time (sec)	UL-94 Rating
A	0.0	30	116	2.8	25	V-0
1	0.5	30	107	3.0	26	V-0
2	1.0	30	106	3.0	24	V-0
3	1.5	30	103	2.9	25	V-0

5

**EXAMPLE 4**

Four LCP compositions were molded, three containing low molecular weight PE, and Comparative Example B not containing PE. The same LCP, PE, and glass was used as in Examples 1-3. The talc used was Supra® Talc (Cyprus Minerals), and the carbon black used was Velvetine® carbon black. The compositions are given in Table 2, where the weight percents are based on the totals of all components of the compositions.

Electrical connectors were injection molded of each sample. Sample B had a minimum cycle time of 19 sec, and the part was deformed by the ejector pin when ejected. Samples 4A and 4B both had minimum cycle times of 14.9 sec, and had no deformation from the ejector pin. Sample 4C had a minimum cycle time of 15.9 sec and showed no deformation caused by the ejector pin. These particular results illustrate the improved mold release properties when PE was present.

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**TABLE 2**

Sample	% LCP	% PE	% Fiber- glass	% Carbon Black	% Talc
B	70	0	30	0	0
4A	69.5	0.5	30	0	0
4B	69	1.0	30	0	0
4C <sup>a</sup>	61.5	0.5	23.3	1.3	13.3

- 5     <sup>a</sup> Made by pellet blending 1 part by weight of a sample containing 68.5% LCP, 1.5% polyethylene and 30% glass fiber, and 2 parts by weight of a sample containing 58% LCP, 20% glass fiber, 20% talc and 2.0% carbon black.

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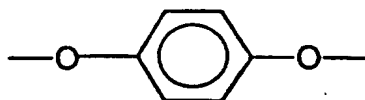
**CLAIMS**

1. A composition comprising an aromatic liquid crystalline polymer and from about 0.05 to about 2.5 percent by weight of said aromatic liquid crystalline polymer of polyethylene, said polyethylene having a molecular weight within the range between about 1,000 to about 10,000.
2. The composition as recited in Claim 1 wherein said polyethylene has a molecular weight within the range between about 1,500 to about 4,000.
3. The composition as recited in Claim 1 wherein said polyethylene is polar.
4. The composition as recited in Claim 3 wherein said polyethylene is made by low pressure polymerization.
5. The composition as recited in Claim 1 wherein the amount of said polyethylene is about 0.1 to 2.0 percent by weight of said aromatic liquid crystalline polymer.
6. The composition as recited in Claim 4 wherein the amount of said polyethylene is about 0.2 to 1.0 percent by weight of said aromatic liquid crystalline polymer.
7. The composition as recited in Claim 1 wherein said aromatic liquid crystalline polymer contains repeat units derived from one or more of 4-hydroxybenzoic acid, 6-hydroxynapthoic acid, terephthalic acid, isophthalic acid, 2,6-naphthalene dicarboxylic acid, 4,4'-bibenzoic acid, hydroquinone, chlorohydroquinone, methylhydroquinone, t-butylhydroquinone, phenylhydroquinone, phenethylhydroquinone, 4,4-biphenol, resorcinol, or 2,6-dihydroxynaphthalene.
8. The composition as recited in Claim 6 wherein said aromatic liquid crystalline polymers contains repeat units derived from one or more of 4-hydroxybenzoic acid, 6-hydroxynapthoic acid, terephthalic acid, isophthalic acid, 2,6-naphthalene dicarboxylic acid, 4,4'-bibenzoic acid, hydroquinone, chlorohydroquinone, methylhydroquinone, t-butylhydroquinone, phenylhydroquinone, phenethylhydroquinone, 4,4-biphenol, resorcinol, or 2,6-dihydroxynaphthalene.
9. The composition as recited in Claim 1 wherein said aromatic liquid crystalline polymer consists essentially of the repeat units:

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- 11 -

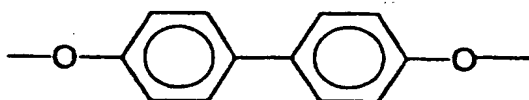
(a) structure (I),



(I)

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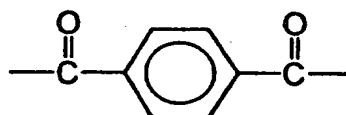
(b) structure (II),



(II)

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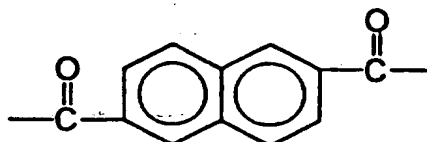
(c) structure (III),



(III)

15

(d) structure (IV),

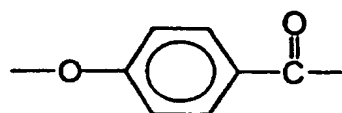


(IV)

and

20

(e) structure (V)



(V)

25 wherein the molar ratio of (I):(II) ranges from 0:100 to 100:0, wherein the molar ratio of (III):(IV) ranges from 85:15 to 15:85, wherein the molar ratio

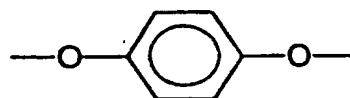
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- 12 -

of the total of (I) and (II) to the total of (III) and (IV) is substantially 1:1, and further wherein there are 100 to 600 moles of (V) per 100 moles of (I) plus (II);

10. The composition as recited in Claim 1 wherein said aromatic liquid crystalline polymer consists essentially of the repeat units:

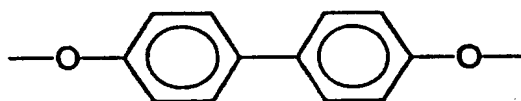
(a) structure (VI)



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(VI)

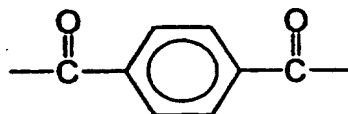
and  
structure (VII)



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(VII)

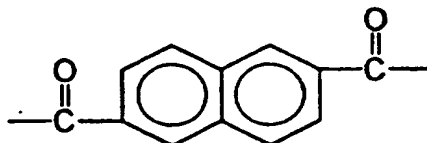
(b) structure (VIII)



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(VIII)

(c) structure (IX)



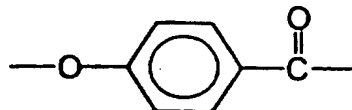
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- 13 -

(d) structure (X)

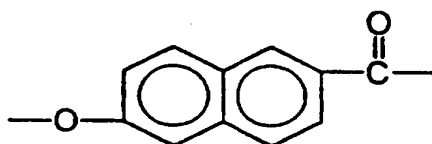


(X)

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and

(e) structure (XI)



(XI)

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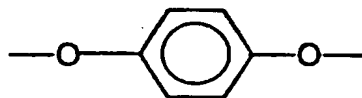
wherein the molar ratio of (VI):(VII) ranges from 0:100 to 100:0, the molar ratio of (VIII):(IX) ranges from 25:75 to 90:10, the molar ratio of the sum of (VI) and (VII) to the sum of (VIII) and (IX) is substantially 1:1, the molar ratio of (IX):(X) ranges from 97:3 to 50:50, and the number of moles of (IX) plus (X) ranges from 100 to 600 per moles of (VI) plus (VII).

15

11. The composition as recited in Claim 6 wherein said aromatic liquid crystalline polymer consists essentially of the repeat units:

(a) structure (I),

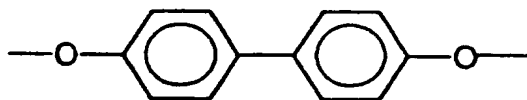
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(I)

(b) structure (II),

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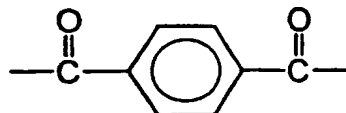


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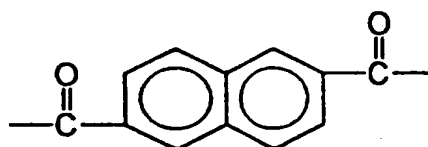
(c) structure (III),



(III)

5

(d) structure (IV),

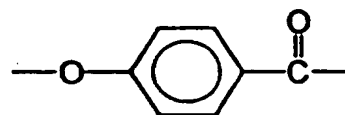


(IV)

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and

(e) structure (V)



(V)

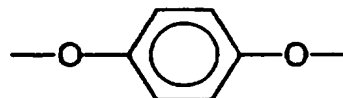
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wherein the molar ratio of (I):(II) ranges from 0:100 to 100:0, wherein the molar ratio of (III):(IV) ranges from 85:15 to 15:85, wherein the molar ratio of the total of (I) and (II) to the total of (III) and (IV) is substantially 1:1, and further wherein there are 100 to 600 moles of (V) per 100 moles of (I) plus (II);

20

12. The composition as recited in Claim 6 wherein said aromatic liquid crystalline polymer consists essentially of the repeat units:

(a) structure (VI)



(VI)

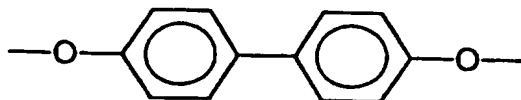
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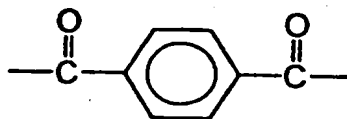
and  
structure (VII)



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(VII)

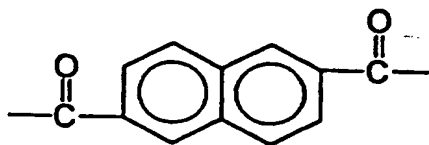
(b) structure (VIII)



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(VIII)

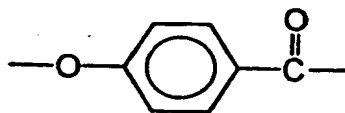
(c) structure (IX)



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(IX)

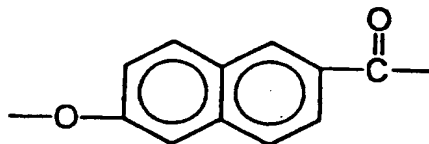
(d) structure (X)



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(X)

and  
(e) structure (XI)



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(XI)

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- 16 -

wherein the molar ratio of (VI):(VII) ranges from 0:100 to 100:0, the molar ratio of (VIII):(IX) ranges from 25:75 to 90:10, the molar ratio of the sum of (VI) and (VII) to the sum of (VIII) and (IX) is substantially 1:1, the molar ratio of (IX):(X) ranges from 97:3 to 50:50, and the number of moles of (IX) plus (X) ranges from 100 to 600 per moles of (VI) plus (VII).

13. The composition as recited in Claim 1 wherein said aromatic liquid crystalline polymer is selected from the group consisting of a polyester, a poly(imide-ester) and a poly(amide-ester).

14. The composition as recited in Claim 1 further comprising glass fiber.

15. The composition as recited in Claim 1 further comprising a filler selected from the group consisting of glass fiber, carbon black, titanium dioxide, talc and combinations thereof.

16. The composition as recited in Claim 1 wherein said aromatic liquid crystalline polymer is a polyester.

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## AMENDED CLAIMS

[received by the International Bureau on 14 February 1996 (14.02.96);  
original claims 1 and 2 amended;  
remaining claims unchanged (1 page)]

1. A composition comprising an aromatic liquid crystalline polymer and from 0.05 to 2.5 percent by weight of said aromatic liquid crystalline polymer of polyethylene, said polyethylene having a  
5 molecular weight within the range between 1,000 to 10,000.
2. The composition as recited in Claim 1 wherein said polyethylene has a molecular weight within the range between 1,500 to 4,000.
3. The composition as recited in Claim 1 wherein said  
10 polyethylene is polar.
4. The composition as recited in Claim 3 wherein said polyethylene is made by low pressure polymerization.
5. The composition as recited in Claim 1 wherein the amount of said polyethylene is about 0.1 to 2.0 percent by weight of said aromatic  
15 liquid crystalline polymer.
6. The composition as recited in Claim 4 wherein the amount of said polyethylene is about 0.2 to 1.0 percent by weight of said aromatic liquid crystalline polymer.
7. The composition as recited in Claim 1 wherein said  
20 aromatic liquid crystalline polymer contains repeat units derived from one or more of 4-hydroxybenzoic acid, 6-hydroxynaphthoic acid, terephthalic acid, isophthalic acid, 2,6-naphthalene dicarboxylic acid, 4,4'-bibenzoic acid, hydroquinone, chlorohydroquinone, methylhydroquinone, t-butylhydroquinone, phenylhydroquinone,  
25 phenethylhydroquinone, 4,4-biphenol, resorcinol, or 2,6-dihydroxynaphthalene.
8. The composition as recited in Claim 6 wherein said aromatic liquid crystalline polymers contains repeat units derived from one or more of 4-hydroxybenzoic acid, 6-hydroxynaphthoic acid,  
30 terephthalic acid, isophthalic acid, 2,6-naphthalene dicarboxylic acid, 4,4'-bibenzoic acid, hydroquinone, chlorohydroquinone, methylhydroquinone, t-butylhydroquinone, phenylhydroquinone, phenethylhydroquinone, 4,4-biphenol, resorcinol, or 2,6-dihydroxynaphthalene.
9. The composition as recited in Claim 1 wherein said  
35 aromatic liquid crystalline polymer consists essentially of the repeat units:

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## INTERNATIONAL SEARCH REPORT

Inv. onal Application No

PCI/US 95/08969

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08L67/00 C08L67/04 C08L77/12 C08L79/08

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP,A,0 462 844 (TORAY INDUSTRIES, INC) 27 December 1991 see page 6, line 40 - line 46; claims 1-3,13-16	1,2,5-16
X	US,A,3 647 920 (R. R. DI LEONE) 7 March 1972 see column 2, line 29 - line 53; claims 1-10	1-4, 13-15
X	DATABASE WPI Week 9002 Derwent Publications Ltd., London, GB; AN 90-011669 & JP,A,00 129 057 (UENO PHARMACEUTICAL) , 24 November 1989 see abstract	1,2,5-16

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

## \* Special categories of cited documents:

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- \*&\* document member of the same patent family

Date of the actual completion of the international search

27 November 1995

Date of mailing of the international search report

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Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax (+31-70) 340-3016

Authorized officer

Decocker, L

# INTERNATIONAL SEARCH REPORT

Int'l Patent Application No  
PCT/US 95/08969

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>CHEMICAL ABSTRACTS, vol. 119, no. 10, 6 September 1993 Columbus, Ohio, US; abstract no. 99421, IVANOV, V. E. ET AL 'Structurization of lubricants by liquid crystalline compounds' see abstract &amp; KHIM. TEHKNOL. TOPL. MASEL, vol. 2 , 1993 pages 17-18,</p> <p style="text-align: center;">---</p>	1-16
A	<p>EP,A,0 111 179 (GENERAL ELECTRIC COMPANY) 20 June 1984 see claims 1-16,20-25</p> <p style="text-align: center;">-----</p>	1-16

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# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 95/08969

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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US-A-3647920	07-03-72	NONE	
EP-A-111179	20-06-84	US-A- 4564658 AU-B- 569266 AU-B- 2160183 CA-A- 1263788 DE-A- 3373821 JP-B- 4075259 JP-A- 59131645	14-01-86 28-01-88 31-05-84 05-12-89 29-10-87 30-11-92 28-07-84

Form PCT/ISA/210 (patent family annex) (July 1992)